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Positron emission intensity in the decay of ^{120g}I†

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Summary. The β^+/EC ratio in the decay of ^{120g}I ($T_{1/2}$ = 1.35 h), i.e. the β^+ emission intensity, was measured for the first time using a high purity ^{120g}I source, which was produced by irradiation of 99.0% enriched ^{120}Te with 15 MeV protons, followed by a chemical separation of radioiodine. Both γ-ray and X-ray spectroscopy were applied. In the former case a comparison of the intensity of the annihilation radiation with that of the 1523 keV γ-ray of ^{120g}I was done and in the latter case with that of the K_α or K_β X-ray of the daughter tellurium. Corrections for very small contributions of ^{121}I and ^{120m}I impurities were evaluated. The literature values for the I_{β^+} of ^{120g}I range between 39 and 81%. Our measurements lead to a value of 56 ± 3%.

Introduction

For quantitation of ^{123}I-labelled SPECT-radiopharmaceuticals the analogue radioisotope ^{120g}I ($T_{1/2}$ = 1.35 h; E_{β^+} = 4.0 MeV; $I_{\beta^+} \geq 39\%$) appears to be very promising since positron emission tomography (PET) can be used. Recent PET-phantom measurements have shown that, despite its high β^+ end-point energy, ^{120g}I is suitable for PET studies [1]. There is, however, some discrepancy regarding the intensity of emitted positrons. In view of the PET application of ^{120g}I, the β^+ emission intensity in its decay needs to be accurately determined.

Early investigations on the characterization of light mass radioisotopes of iodine involved mass separation of spallation products [cf. 2, 3]. The first unambiguous identification of ^{120g}I was achieved radiochemically via the ^{127}I(p,8n)^{120}Xe $\xrightarrow[40\,\text{min}]{\beta^+,\text{EC}}$ ^{120g}I route in irradiations of NaI with 340 MeV protons [4]. The half-life and two β^+ groups with end-point energies of 2.1 and 4.0 MeV were characterized. However, the β^+/EC decay ratio could not be estimated with certainty, since the radioiodine obtained via the decay of radioxenon contained ^{120g}I, ^{121}I and ^{123}I in admixture [4]. Later decay scheme investigations on highly neutron deficient radioisotopes of iodine involved direct production of radioiodine via high energy reactions, for example the (p,xn) process on tellurium at $E_p \leq 150$ MeV [5], (γ,7n) reaction on ^{127}I at 830 MeV [6], (e,e′7n) and (π,π′7n) reactions on ^{127}I [7, 8], (p,spall) process on La [9, 10], (γ,spall) process on ^{133}Cs [11] and heavy-ion induced reactions on Ag, Pd, Cd and ^{238}U [cf. 12, 13]. In the case of direct production routes, an isomeric state in mass 120, i.e. ^{120m}I ($T_{1/2}$ = 53 min) was also observed [cf. 12]. In each case the radionuclide ^{120g}I contained appreciable quantities of isotopic impurities.

The intensities of γ-rays associated with the decay of both ^{120m}I and ^{120g}I have been determined rather well [cf. 9]. However, the β^+/EC decay ratio has been and is still the subject of controversy. In one measurement [3] the β^+ intensity of ^{120g}I was reported as 46 ± 3%; in another experiment [14] the amount of β^+ emission relative to total γ-emission was deduced as 39 ± 13%. In one compilation [15] the intensity of β^+ emission has been listed as 81%. Since all studies to date were performed using impure radioiodine, the resulting β^+/EC ratio for ^{120g}I is rather uncertain. The present work attempts to determine the β^+ intensity by measurements on a very pure ^{120g}I sample.

Experimental

Preparation of ^{120g}I source

Thin sample of 99.0% enriched ^{120}Te was prepared by electrolytic deposition on a 20 μm titanium foil and covered tightly with a 10 μm Al foil [16]. Three such samples were prepared and each was irradiated at the compact cyclotron CV 28 of the Forschungszentrum Jülich (FZJ) for 30 min with 15 MeV protons at a beam current of 100 nA. After irradiation the Al cover was removed and the ^{120}Te layer was dissolved in 8 mL aqua regia. The solution was diluted with 20 mL water and the ^{120}Te was recovered via reduction with hydrazine and filtration. A few mL of concentrated ammonia was then added to the filtrate to get a pH value above 9. After adding 1 mL of 0.1 M NaI carrier solution, 2 mL of 0.2 M $Na_2S_2O_3$ and 1.5 mL 0.1 M $AgNO_3$ solution, the radioiodine was precipitated as AgI and filtered. Thereafter it was dried, the filter carrying the precipitate was mounted on a thin polyethylene foil, covered by another polyethylene foil, and subjected to counting.

Measurements

For determination of the β^+/EC decay ratio, both γ-ray spectroscopy and X-ray spectroscopy were applied. For the former a high-resolution HPGe detector was used. The efficiency of the detector was determined as reported earlier

† Dedicated to the memory of Professor Alfred P. Wolf.

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Table 1. γ-ray intensities in the decay of iodine isotopes encountered in this work*.

Nuclide	Half-life	E_γ [keV]	I_γ [%]
^{120m}I	53 min	1345.9	18.9
		560.4	100
		601.1	87.0
^{120g}I	1.35 h	1523.0	11.2
		560.4	73
		601.1	5.8
		641.1	9.1
^{121}I	2.12 h	212.2	84.3

*: Data taken from ref. [17].

Table 2. Intensities of tellurium X-rays in the EC decay of iodine isotopes*.

X-ray	Energy [keV]	Intensity [%]
K_{α_1}	27.47	46.2
K_{α_2}	27.20	25.0
K_{β_1}	30.98	8.21
K_{β_2}	31.88	2.37
K_{β_3}	31.94	4.26

*: Data taken from ref. [17]. Intensities refer to 100 EC events, cf. page F-45.

(cf. ref. [16]). For complete annihilation of the high energy positrons, the ^{120g}I source was placed between two copper plates, each 4 mm thick. The radionuclidic composition of the source was determined by counting without copper absorbers and estimating the absolute activities of the expected radioisotopes. The absolute activities of ^{120g}I and ^{121}I were determined via the 1523 keV (11.2%) and 212.2 keV (84.3%) γ-rays, respectively. For ^{120g}I, the other stronger γ-rays at 560 and 641 keV were not employed due to some contribution from the ^{120m}I. The ^{120m}I activity, on the other hand, was determined by measurement of the 601.1 keV (87%) γ-ray after subtraction of the ^{120g}I contribution (5.8%) at the same energy. The γ-ray intensities and other decay data were taken from ref. [17] and are summarized in Table 1. At the start of measurement, the relative amounts of the three activities were found to be: ^{120g}I (100), ^{120m}I (2.4) and ^{121}I (1.4). The radioisotope ^{121}I was produced via the (p,2n) reaction on the small amount of ^{122}Te present in the enriched ^{120}Te sample. The ^{120g}I sample was thus very pure. Nonetheless, the small amounts of the impurities present needed to be taken into account while calculating the β^+/EC decay ratio.

For X-ray counting a Si(Li) detector with an energy resolution of 45 eV at 14.4 keV was used. The emphasis was on the measurement of K_α and K_β X-rays of Te. In the former case a sum of the $K_{\alpha 1}$ and $K_{\alpha 2}$ lines (summed intensity 71.2%) was determined and in the latter a sum of the $K_{\beta 1}$, $K_{\beta 2}$ and $K_{\beta 3}$ lines (summed intensity 14.8%). The intensities of the individual lines (as percentage of EC decay) were taken from ref. [17]; a summary is given in Table 2.

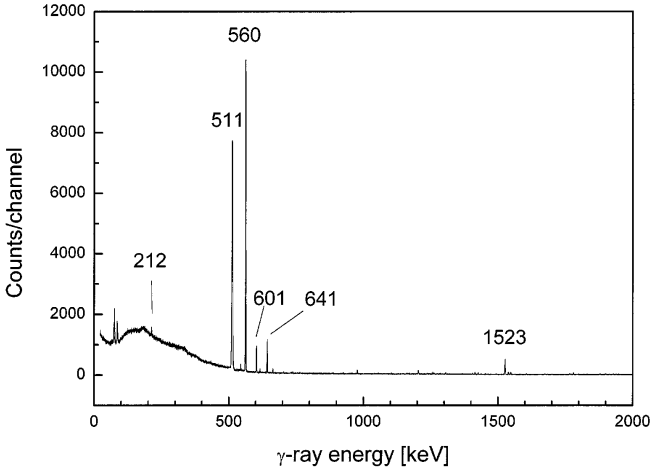

Fig. 1. HPGe detector γ-ray spectrum of radioiodine. All the marked γ-lines (except the one at 212 keV) are attributable to ^{120g}I. The weak 212 keV γ-ray is due to ^{121}I.

Calculation of β^+/EC decay ratio

For calculation of the β^+ emission intensity of ^{120g}I via the γ-ray spectroscopic measurement, half the peak area around the 511 keV annihilation radiation, corrected for the efficiency of the detector, was divided by the absolute activity of ^{120g}I (see above). The contribution to the 511 keV peak area from the positron emission of ^{120m}I could not be subtracted, because the β^+/EC decay ratio also in the case of ^{120m}I is unknown. This, however, should not cause much uncertainty since the contribution of ^{120m}I was anyway small (see above).

For calculation of the β^+/EC decay ratio via X-ray spectroscopy, and therefrom the β^+ emission intensity (I_{β^+}) of ^{120g}I, the following formula was used:

$$I_{\beta^+} = \frac{0.5 \cdot A_{511} \cdot \varepsilon_{511}}{0.5 \cdot A_{511} \cdot \varepsilon_{511} + A_{\text{X-ray}} \cdot \varepsilon_{\text{X-ray}} \cdot 1/I_{\text{X-ray}}} \cdot 100$$

with

A_{511} = Peak area of 511 keV annihilation radiation.
$A_{\text{X-ray}}$ = Peak area of K_α or K_β keV X-ray radiation.
ε_{511}, $\varepsilon_{\text{X-ray}}$ = Efficiency of detector at respective energies.
$I_{\text{X-ray}}$ = X-ray intensity relative to EC.

Both A_{511} and $A_{\text{X-ray}}$ were corrected for decay.

Results and discussion

The results of γ-ray and X-ray spectroscopic measurements are given in Figs. 1 and 2, respectively. Evidently only iodine radioisotopes are present and the dominant contribution is from ^{120g}I. For an unambiguous assignment of the 1523 and 511 keV γ-radiation and the 27 and 31 keV X-rays to ^{120g}I decay, the half-lives were checked. Typical results are given in Fig. 3. The result for the 1523 keV γ-line was 81.9 ± 2 min and agreed excellently with the half-life value of ^{120g}I given in the literature as 81.0 ± 0.6 min. The half-life via the 511 keV radiation emerges as 81.5 ± 2 min and confirms the purity of ^{120g}I, i.e. there is no significant contamination from any other positron emitter (the contribution of ^{120m}I was too low to measure). The positron emission

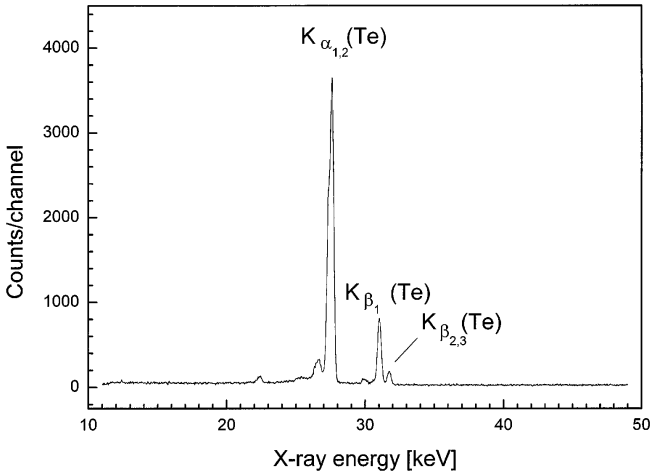

Fig. 2. Si(Li) detector X-ray spectrum of radioiodine (mainly ^{120g}I).

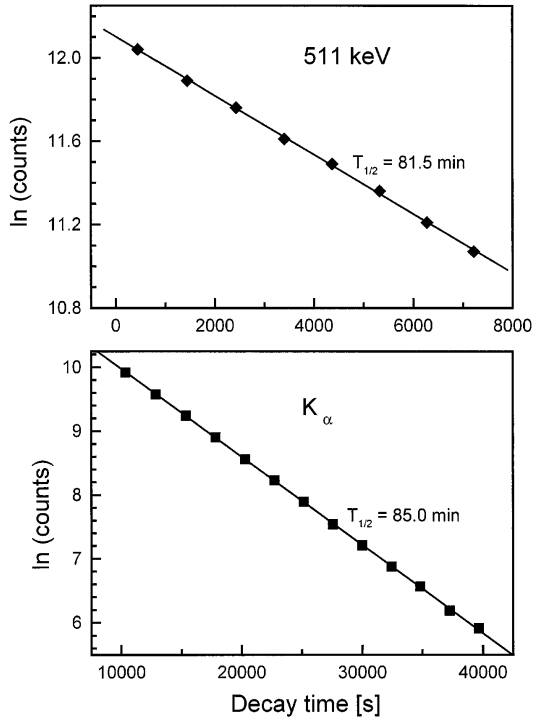

Fig. 3. Decrease in intensity of 511 keV annihilation radiation and 27 keV K_α X-radiation of a radioiodine sample as a function of time.

intensity for ^{120g}I deduced from several independent β^+/γ measurements was found to lie between 54.4 and 60.4% with a mean value of 58.4%. The uncertainties are given in Table 3. They were estimated considering the relative uncertainties in the efficiency of the detector (2%) and the peak area analysis (3%). The higher negative error is due to the 3% uncertainty in the contribution of ^{120m}I.

The half-life measurement via K_α and K_β X-rays gave a value of 85 ± 0.4 min (cf. Fig. 3). The somewhat higher value is due to the presence of some ^{121}I which decays 88% [cf. 17] via EC. Adopting the contribution of ^{121}I ($T_{1/2}$ = 2.12 h) in ^{120g}I as 1.4% (see above) and taking into account the uncertainties in the efficiencies of the detectors and the peak areas used, the half-life value of 85 min appears reasonable. With an uncertainty of 5% in the detector efficiency and 8% in the peak area, due to the contributions of ^{121}I and ^{120m}I, the positron emission intensity of ^{120g}I amounts to 52.6% via K_α and 52.8% via K_β radiation. The uncertainties are given in Table 3.

Table 3. β^+ branching in the decay of ^{120g}I.

Method of measurement	I_{β^+} (%)
γ-ray spectroscopy	$58.4^{+2.0}_{-4.0}$
X-ray spectroscopy using K_α (Te)	$52.6^{+4.7}_{-1.6}$
X-ray spectroscopy using K_β (Te)	$52.8^{+5.2}_{-2.3}$
Weighted average	56.1 ± 3.2

It should be mentioned that in the case of X-ray counting the uncertainties are somewhat larger since, on the one hand, absolute uncertainties in the efficiencies of the two detectors were involved and, on the other, the ^{121}I contribution had to be taken into account. We therefore gave a slightly higher weight to γ-ray spectroscopic measurements (60%) as compared to X-ray measurements (40%). Thus, using a weighting factor of (3:1:1) to the three measurements listed in Table 3, a value of 56.1 ± 3.2% for I_{β^+} of ^{120g}I was obtained.

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